

Soot formation in turbulent nonpremixed flames

Soot formation in turbulent nonpremixed flames is an important process in many practical combustion environments. In diesel engines, soot emission is a pollutant with adverse health impacts, and its formation reduces combustion efficiency through incomplete combustion of fuel. Soot is responsible for a flame's luminosity, and the bulk of radiative heat transfer in flames and fires is due to radiative emission from soot. In fires, substantial quantities of soot (smoke) are emitted that can result in radiative shielding of surrounding objects, such as buildings or explosive containers. Obtaining an understanding of the fundamental processes governing soot formation and its interaction with flames is crucial for designing equipment, such as engines, gas turbines, and boilers, and to accurately predict fire hazards.

The formation of soot in flames is a complicated process involving hundreds of chemical species

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Fluorescence resonance energy transfer in single quantum dot-dye hybrids

Fluorescence based probes can be used to sense many properties in complex chemical and biological systems, such as pH, ion concentration, and ligand binding. Fluorescence resonance energy transfer (FRET) between donor and acceptor fluorophores is a sensitive detection mechanism for these probes if the separation between the fluorophores can be designed to depend on the quantity of interest. As donor fluorophores, semiconductor quantum dots (QDs) offer significant advantages over fluorescent dyes, including superior photostability, broad absorption, and narrow, size-tunable emission. Thus, there is rapidly growing interest in QD-organic dye hybrids for FRET-based sensing. [1-4] To probe nanostructured environments,

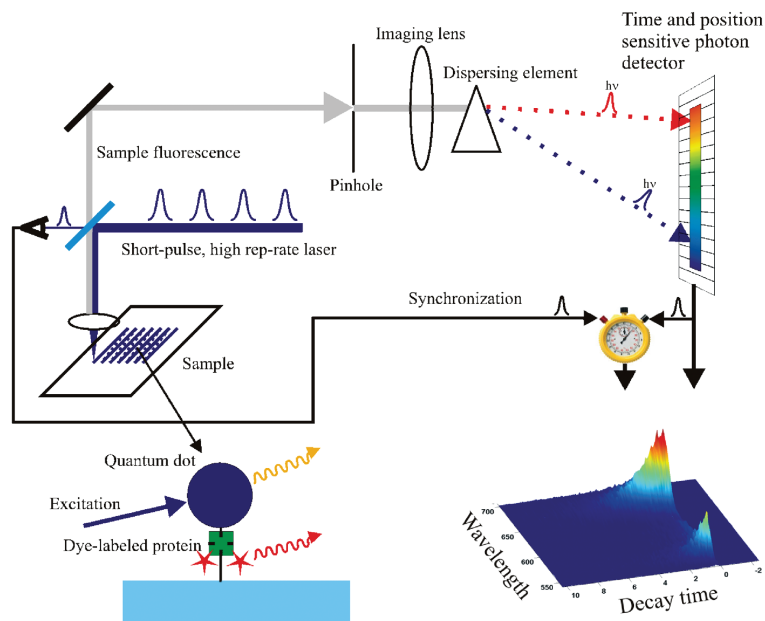


Figure 1. Experimental setup. In our time-resolved, multi-spectral microscope, a high repetition rate pulsed laser excites the sample. Fluorescence is collected and directed onto a time-and-position sensitive detector. The output of this detector referenced to the excitation pulses gives the fully correlated fluorescence spectrum and time decay. A schematic of the quantum dot-dye hybrid used in these experiments is at lower left.

it is desirable to have sensors that will operate at the single particle level. For most purposes,

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Mark Linne presents workshop for Post Docs: "How to Find a Faculty Position"

On June 12, Mark Linne, Sandia Manager, Combustion Chemistry Department, and former Professor, Engineering Division, Colorado School of Mines (1989), and Professor, Combustion Physics Department, Lund Institute of Technology, Sweden (2002), presented a workshop to help post-doctoral employees at Sandia learn about the academic faculty hiring process. Dr. Linne is well-qualified to speak on this topic as he served on thirteen faculty search committees and chaired six.

Dr. Linne presented practical advice and insider information on the following topics: (1) How to prepare for the job search; (2) Warnings and conventional wisdom; (3) How to find and target an open position; (4) The search and hiring process from the inside; (5) How

to apply; (6) How to interview; (7) What can happen after the interview; and (8) What to do if offered a position.

How to Prepare for the Job Search

When beginning the process of searching for a faculty position, prospective candidates should bear in mind that they must interact with students. They must want to teach and advise students, and they must seek funding for the university and their department. This is an ongoing activity. They must also endure the tenure process, and may find that no matter how good they are at their job, or how conscientiously they carry out all required duties, they may not in fact be granted tenure. State-funded educational institutions are

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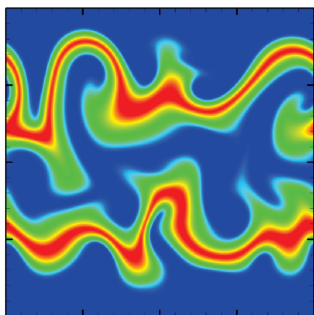


Figure 1. Temperature contours at 1.75 ms. Scale is 300 K (blue) to 2100 K (red).

turn depends on the relative convection between the soot and flame, as well as the residence time of the soot in reactive regions.

The relative simplicity of laminar flames lends itself to detailed experimental and numerical investigation. However, limited progress has been made in turbulent environments because of the complexity of the unsteady flow field, the large scale separation between the bulk flow, and the smallest flame and soot scales. Experimental data in turbulent sooting flames is usually limited to mean quantities, with no information about the detailed soot-flame-turbulence structure. Jackie Chen, Distinguished Member of the Technical Staff at Sandia, and David Lignell, a PhD student in the Chemical Engineering Department at the University of Utah, have performed the first direct numerical simulations (DNS) with detailed chemistry and transport of 3-D turbulent flames with soot formation. DNS resolves all length and time scales of the velocity field and chemical composition of the combustion environment, unlike RANS and LES approaches, which rely on subgrid models to account for unresolved turbulence-chemistry interactions. The goals of this research are two-fold: (1) to provide fundamental insight into the physical processes at work, and (2) to provide high fidelity data for evaluation and development of advanced combustion models.

Figure 1 shows temperature contours of a two-dimensional simulation. [1] A slab of pure ethylene is sandwiched between air streams overlaid with an isotropic turbulent field. Flame sheets exist at the fuel/air interface, which are wrinkled by the turbulence. A validated, reduced ethylene mechanism consisting of 19 transported and 10 quasi-steady-state species and 167 chemical reactions describes the combustion environment. [1] A semi-empirical, 4-step soot mechanism is employed that accounts for nucleation, growth, oxidation, and coagulation. The first two mass moments of the soot particle size distribution are transported, which correspond to the soot mass fraction and number density. The domain size is 2 by 3 cm in the horizontal and vertical directions, respectively. The simulations were performed on the Thunderbird cluster at Sandia National Laboratories.

The defining characteristic of turbulent combustion is the unsteady, multi-dimensional structure of the flames, most notably indicated by

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which grow and combine to form soot particles. Soot is formed on the fuel-rich side of nonpremixed flames, and, as a particulate phase, behaves differently than the surrounding gaseous species, especially in its transport properties. The concentration of soot and its proximity to the high temperature burning flame zone strongly impact the reaction rate of the soot: both formation in fuel-rich zones and destruction in oxidizing regions. This soot-flame proximity in

turn depends on the relative convection between the soot and flame, as well as the residence time of the soot in reactive regions. flame curvature, as evident in Figure 1. While the gaseous species are only moderately affected by flame curvature in the present simulation, the soot concentrations are remarkably sensitive to flame curvature. We have found that soot concentrations peak where the flame has its center of curvature in the fuel stream (negative curvature). This effect is not due to thermal focusing of the flame, as might be expected, but rather is due to differential diffusion of the soot relative to the flame. As a particle phase, soot is mainly convected with the fluid, with a secondary thermophoretic diffusion velocity. In regions of negative flame curvature, the soot is convected towards the flame zone where it is at a higher temperature and has a higher reactivity. The relative motion between the soot and the flame is quantified using the motion of isocontours of the stoichiometric mixture fraction, relative to convection. Figure 2 is a contour plot with the stoichiometric surface colored by this relative velocity, along with soot contours in black and white, and fluid velocity vectors. This plot shows that high levels of soot correspond to high values of the relative velocity, where soot is convected into the flame. In these regions, the radiative soot emission is as much as 15 times higher than in regions of negative flame velocity where soot is convected away from the flame zone.

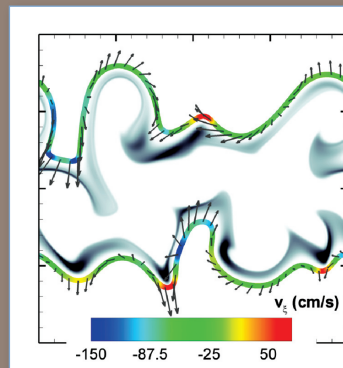


Figure 2. Soot mass fraction contours (dark is peak at 0.002) at 1.75 ms with stoichiometric surface colored by the velocity of the surface relative to fluid convection. Fluid velocity vectors also shown.

The quantification and modeling of the relative motion between soot and the flame (or soot and mixture fraction in general) is of fundamental importance because this differential mixing process impacts radiation heat transfer rates, soot concentrations, and may lead to soot-flame breakthrough and flame quenching. To better understand and quantify these processes, Sandia researchers David Lignell and Jackie Chen have performed three-dimensional simulations similar to the two-dimensional simulation, but in a temporal jet configuration. [2] Figure 3 shows temperature and soot mass fraction contours. These simulations were performed with a grid of 228 million computational nodes, on the Red-storm supercomputer at Sandia National Laboratories. In these simulations, the combined effects of differential transport of soot in the mixture fraction coordinate compete with the mixout of the fuel core as the global soot is first transported towards

fuel-rich regions, then back towards the fuel-lean oxidizing regions. The simulation results are being used for *a priori* testing of combustion models, such as conditional moment closure modeling of soot formation. Future simulation efforts will include more complex soot particle models and chemical mechanisms to gain further insights into sooting reaction-turbulence interactions.

References

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2007

SUMMER PSI INTERNS ENGAGE IN COMBUSTION PROJECTS AT CRF

S u m m e r I n t e r n s

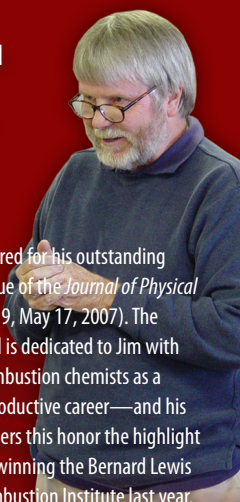
Summer interns in the Physical Sciences Institute (PSI) program work with CRF staff mentors on various combustion research projects. PSI interns come from across the nation, from high schools, universities, and post graduate programs. Melissa Harmon, from Manteca East Union High, is working with Melanie Steadman as an administrative assistant. Victoria Lee, a returning intern from California Polytechnic State University, San Luis Obispo, is continuing to work with Joe Oefelein on developing advanced preconditioning methods for reacting flows. Tom Goldstein, a graduate student in applied mathematics at UCLA, and Steve Widmer, a PhD candidate in mathematics at the University of North Texas, are both working with Habib Najm, and are respectively co-mentored by Youssef Marzouk and Bert Debusschere. Steve is working on code development for uncertainty quantification in chemical systems using multiwavelet constructions. Tom is working on Bayesian methods for data assimilation and construction of uncertain flame models from empirical measurements. Paul Abel, a chemical engineering major at MIT, is working with Craig Taatjes on the kinetics of propyl and ethyl radicals reacting with oxygen. Spencer Behling, from Brigham Young University, is working with Nils Hansen to characterize temperature profiles for low pressure pre-mixed laminar methane flames. Hem Wadhar, a mathematics and physics major from UCLA, is working with Tom Settersten on developing a software package for laser induced fluorescence. Mike Starr, a sophomore in physics at Harvey Mudd College, is also a returning intern, working with David Osborne on spectroscopic quantum measurements of gas phase reactions. Matthew Fife, a graduate student at UC Davis majoring in mechanical and aeronautical engineering, is working with Paul Miles on diesel engine design stress models. Ryan Gehmlich, a senior in mechanical engineering at UC Davis, is a third year intern at Sandia, working with Lyle Pickett on the calibration of fuel injectors. John Bustamante, a senior in mechanical engineering and mathematics at Marquette University, is working with Dick Steeper to develop a calibration device for a sonic flow nozzle. Kyle Kattke, a senior majoring in mechanical engineering at the South Dakota School of Mines and Technology, is working with Mark Musculus to model the injection of diesel spray. And Rob Knaus, a first year graduate student in mechanical engineering at the University of Illinois, is also working with Joe Oefelein on characterizing filtered scalar dissipation of turbulent flows.

Lead mentor of the PSI program is Eilene Cross, who may be contacted by email at ecross@sandia.gov.



Physical Science Institute summer interns—Back row left to right: Paul Abel, Ryan Gehmlich, Matthew Fife, John Bustamante, Kyle Kattke, Mike Starr, and Steve Widmer. Front row left to right: Spencer Behling, Victoria Lee, Tom Goldstein, Rob Knaus, and Hem Wadhar (standing).

Jim Miller honored with Festschrift issue of *Journal of Physical Chemistry*



Jim Miller has been honored for his outstanding career by a Festschrift issue of the *Journal of Physical Chemistry* (Vol. III, Issue 19, May 17, 2007). The entire issue of the journal is dedicated to Jim with articles submitted by combustion chemists as a tribute to his long and productive career—and his 60th birthday. Jim considers this honor the highlight of his career, along with winning the Bernard Lewis Gold Medal from the Combustion Institute last year.

The cover of this issue includes a montage of images that shows the impact of Jim's work in physical chemistry and combustion. A representation of the propargyl radical, whose reactions are key to soot formation in hydrocarbon flames, and images of sooting flames are superimposed on figures taken from several of Jim's numerous publications.

The issue also includes an essay by Jim, titled "My Life and Career (So Far) in Combustion Chemistry." Jim began working at Sandia in the spring of 1974 and helped open the Combustion Research Facility in 1980. Among the many achievements of his distinguished career is the development of CHEMKIN, the *de facto* standard software for modeling chemical kinetics in combustion.

Jim is currently working with Stephen Klippenstein of Argonne National Laboratory on developing and implementing a theoretical apparatus for studying chemical reactions involving multiple, interconnected potential wells. Such reactions are of paramount importance in the formation of aromatic compounds, polycyclic aromatic compounds, and soot in flames of aliphatic (non-cyclic) fuels.

To learn more about Jim's long and influential career, visit the American Chemical Society Publications home page at: <http://www.pubs.acs.org/journals/jpcafh/>.



Fluorescence resonance energy transfer in single quantum dot-dye hybrids (Continued from page 1)

multiple acceptors are bound to a single QD hub that serves as the sole energy donor. A major obstacle to single-particle measurements with these QD-multiple-acceptor systems is that the FRET signal must be distinguished from acceptor dye photobleaching events, flickering emission caused by the QD donor blinking, and direct excitation of the acceptors.

In collaboration with Dr. C. Shan Xu and Prof. Haw Yang at U.C. Berkeley Sandia researchers Carl Hayden and Hahkjoon Kim are studying QD-dye FRET hybrids to understand the interaction between the QDs and dyes and to develop methods for determining statistically significant FRET changes in the presence of interfering signals. We assemble the QD-dye hybrids using biotin conjugated, 605nm QDs, which strongly bind streptavidin proteins labeled with Cy5 dye. Using excitation with 480nm laser pulses at 20MHz, our time-resolved, multi-spectral microscope [5] measures the wavelength, emission delay relative to excitation (excited-state lifetime), and chronological time (intensity) for each fluorescence photon detected from individual hybrid particles. The initially excited QD donor emission at ~605nm is readily distinguished from the acceptor dye emission around 675nm in the spectrally resolved data. The experimental setup is shown in Figure 1.

Figure 2 shows typical data from a single hybrid. At the top are intensity traces of the QD and dye emission. By analyzing these intensity traces using a changepoint method [6] and looking for synchronized changes in the donor and acceptor emissions, we discriminate changes in FRET from QD and dye blinking. The emission spectrum (Figure 2c) shows that initially energy transfer to the dye (FRET) is strong, but after ~5 seconds the FRET and hence red emission disappears due to photobleaching of the acceptor dyes (Figure 2d). The time evolution of the QD and dye spectral components shown at the bottom of Figure 2 directly illustrates the energy transfer process. Before dye photo-bleaching, the donor lifetime is quenched due to energy transfer to the acceptor. Energy transfer to the acceptor is verified by observing the slower rise time of its emission. In addition, the decay of the acceptor emission is prolonged from its ~1.4 ns intrinsic lifetime because energy transfer can occur throughout the longer donor lifetime (Figure 2e). The form of the acceptor emission transient is important proof that the acceptor emission is from FRET and not direct excitation. Once the dyes photobleach, the QD lifetime increases as it is no longer quenched (Figure 2f). The results of this work provide a robust approach for using QD-dye hybrid FRET-based sensors in single particle applications.

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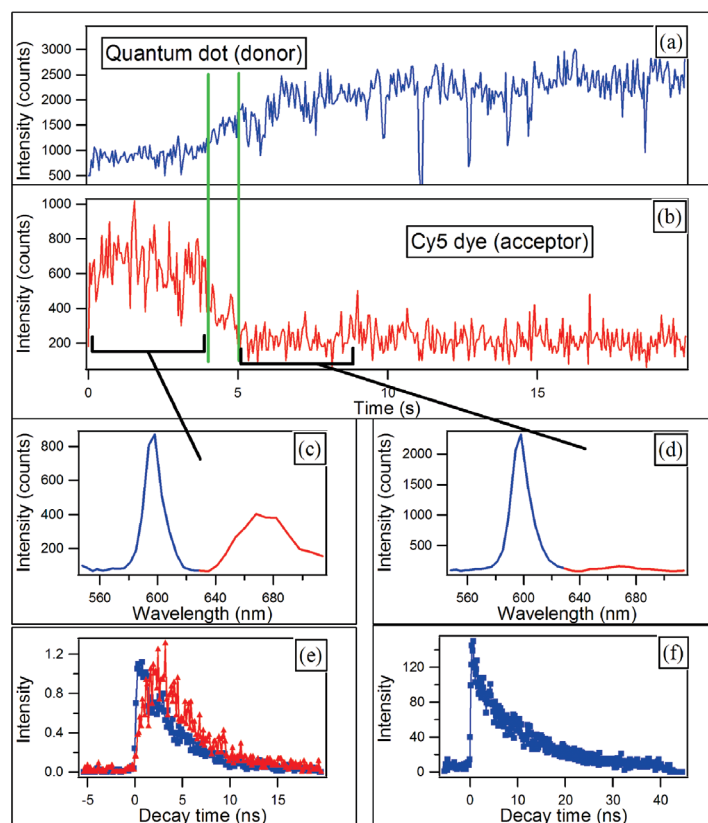


Figure 2. Data from a single QD-dye hybrid. Panels (a) and (b) show intensity traces with time for the QD and dye channels respectively. At short times, the energy transfer is efficient so the QD intensity in (a) is low and the dye intensity (b) is high. This is apparent in the spectra in (c) and (d). The green lines show the photobleaching times for individual dyes identified from our multiparameter statistical analysis. Panel (e) shows the time-resolved energy transfer from initially excited donor (blue) to acceptor (red). The energy transfer from the donor extends the acceptor decay time to match that of the donor. Once the acceptor dyes are photobleached (f), the donor lifetime increases because it is no longer quenched.

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Mark Linne presents workshop on finding a research faculty position (Continued from page 1)

also under increasing pressure from legislators to successfully complete their mission with less money. Candidates may therefore find that they have to work under very restrictive financial constraints, which may affect salary and monies available for capital equipment and supplies. All faculty members must also be prepared to serve on time-consuming committees, which may divert energy and attention from primary duties as professor and educator.

There are, of course, some benefits to weigh in the balance. Tenured faculty members have great freedom—once they have taught their required classes and advised students in their care. In addition, teaching can be very rewarding and may even be fun. A side benefit is that teaching presents new perspectives on the material in ways that may not have been thought about before. The work atmosphere and collegial relationships can also be stimulating. New faculty may also be put in charge of creating a research program with graduate students and post doctoral fellows.

Warnings and Conventional Wisdom

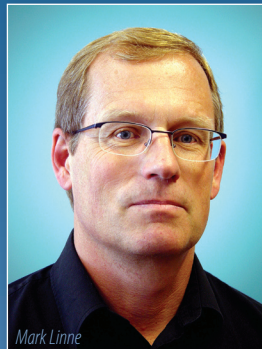
Almost every posted position receives 200 to 300 applicants. The successful candidate's goal is to be among the group of top five candidates. The final selection may be determined by factors out of the candidate's control, like concerns about tenure. Even aspiring to be a great teacher may not matter in the end. Universities are quite conservative and unwilling to take risks. If a candidate steps outside of conventional wisdom, the committee is most likely to think "There must be something wrong with him/her" and hire one of the other top five candidates. Note that only 1% of the faculty in science and engineering at US universities have ever worked in industry or the private sector. Those who ignore conventional wisdom may find it difficult to get back on track.

What is the conventional wisdom that universities and departments apply to the search process? Most deans and department heads believe that there is a pot of gold at the end of the rainbow and if they find the right young star, the pot will then be theirs. Some candidates may claim to be that bright star by presenting themselves as "newly minted" with the following shining attributes: (1) They have successfully identified the latest hot research topic and are already fully engaged in it; (2) They have significant publications in the best journals; (3) They have already demonstrated the ability to find serious research funds; and (4) They are well known and well regarded in the community, and perhaps even a world leader in their field, for which they can provide glowing references and affidavits from the best in the business. All this is of course absurd—that a candidate should already be deserving of tenure when still so young and just starting out. Some candidates, however, manage to convince people that they are such a person, and they therefore self-fulfill their prophecy by high self-confidence that in fact must be justified. Other candidates do succeed, but the question of tenure is always foremost. Tenure is based on the following: (1) Can the candidate do research well?—which means find funding at an absolute minimum of \$200,000 per year, equipping laboratories and facilities, attracting and teaching graduate PhD students, and producing papers (an absolute minimum of two per year after four years); (2) Can the candidate perform service to the university, in professional societies, on campus, and within their department?; (3) Can the candidate teach, even at a minimally acceptable level? In addition to all this, tenure is also a matter of whether or not the candidate fits in with the department, with other faculty members, and with the goals and structure of the department needs within the university.

How to Find and Target an Open Position

To succeed at finding a faculty position, prospective candidates must start their search well in advance of when they expect to start their new career. The best way of finding a position is still through personal connections and at conferences, particularly where the

candidate presents papers. At such occasions, prospective candidates should boldly ask about possible openings. In fact, the more a candidate negotiates a position outside of official channels, the better his or her chances are. They also need to enlist the help of their supervisor and former thesis advisor. They certainly need also to pursue the traditional channels by searching for listings in professional journals, such as *Physics Today*, *Chemical and Engineering News*, *ME Magazine*, *ASEE*, etc. And they must use the Internet and check useful web sites, such as <http://www.higheredjobs.com> and <http://www.academic-careers.com>, and the *Chronicle of Higher Education*, although these sources usually list smaller colleges. The successful candidate searches directly the colleges and other groups of interest and the listed job openings on their web pages. When a position of interest has been found, successful candidates make sure the position closely fits and matches their skills and background. Successful candidates also use strategy—a few carefully crafted applications are better than a lot of poor ones. A good strategy is to find a person on the search committee to talk to, to find out more about the position and form a better match of interests and skills. All prospective candidates seeking faculty positions should be genuinely interested and be certain they get all the information needed for a successful application. This is also a perfect opportunity to alert the people on the inside, as they may become internal advocates.



Successful candidates also find out about the institution itself, about the real situation at the college under consideration—student load, teaching load, general budget, research volume of the various faculty and their areas of specialization. A good technique is to search individual faculty web pages to find out more detailed information. A good resource for data about colleges and universities is <http://www.asee.org/publications/profiles/index.cfm>. It may be that a widely advertised faculty position may not be so desirable when the teaching loads are known. Successful candidates find out about these conditions and only apply for positions that are really a suitable fit for their talents, skills, and career expectations.

The Search and Hiring Process from the Inside

From the inside, the search and hiring process proceeds by different avenues, but when a determination is made that a faculty position needs to be filled, the school and search committee write up a definition of the position. The search committee then meets to set the evaluation criteria. They then write an advertisement based on these criteria, with job requirements meeting point by point the criteria.

How to Apply

Successful candidates make sure, before they apply, that they know what the search committee wants, based on the job description, point by point, and in general. How serious are they about expecting to hire an excellent teacher? Or are other criteria more important? Research and funding capabilities are of course universal. To succeed, the candidate's qualifications must meet the minimum evaluation criteria to convince the search committee this particular candidate satisfies the criteria better than anyone else. The search itself is conducted in stages. First, each member of the committee reads and ranks all the applicants on a form. With 200 to 300 applications to review, they will spend very little time at this stage, so candidates need to be sure that they meet at least the minimum criteria. At this stage, the most resourceful candidates help the committee members put their resumes at the top of the pile by matching criteria point by point, which actually helps the committee members fill in the forms. Another good technique is to include a cover letter with a nice introductory paragraph that succinctly lists all the criteria that they need to satisfy. The committee then meets to compile scores on the applications, with the goal of narrowing the search down to ten serious applications. After this, they each assess and evaluate again the ten applications. The committee then pares the list of ten down to a "short list" of three to five applicants. At this point the head of the

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Mark Linne

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search committee calls the applicants on the short list to let them know they are on the list and ask permission to contact their references. The committee then contacts references, often asking for letters and also for other names from the references to contact. Finally, the committee meets to decide whom to invite for a personal interview.

To prepare for the next stages of the process, successful candidates make certain that they have everything in order. The best resumes are well organized, partitioned into sections that show education and degrees, with thesis title and advisor's name; work experience, including teaching; awards, papers, publications, and presentations. In addition, most successful candidates include a well-defined and relevant 1-2 page research plan and a separate 1 page teaching plan. Remember—Most deans and department heads believe that there is a pot of gold at the end of the rainbow. . . they will hire the candidate they think can deliver what is proposed. Candidates should not, therefore, propose an arcane research topic—it's the kiss of death. They should choose a topic that is current, a hot research topic that is likely to receive viable funding. For this, candidates should get advice from their advisor and also find out who's funding what by searching the Internet, NSF, DOE, AFOSAR, ARO, for example. They also need to show that they understand the funding process, to convince the search committee that indeed they will be able to get funding for this exciting new area of research at their institution.

The teaching plan should show genuine interest in teaching and that the candidate does not plan simply to retire after receiving tenure but also plans to pursue research with fervor throughout their career. Successful candidates therefore

propose to develop a couple of innovative classes (probably at the graduate level) in a new area for the department.

How to Interview


When representatives of the university contact chosen candidates for an interview, they expect them to be prepared to answer all their questions about research, teaching, and any other topics related to the position. The successful candidate, therefore, answers the questions in a credible way that does not scare them off with huge demands. "Sink the hook first." After they have chosen the person they want to hire, it is possible then to make demands that they fill their part of the bargain. The interview process itself takes more than a day. It is exhausting. Candidates are asked to give a seminar, where they are judged on their performance, their ability to teach, to convince, on the nature and seriousness of their subject, if they are well prepared and ready to answer questions, and if they keep to the time limit and on topic. Good candidates prepare in advance and practice their talk so they can give it smoothly and with confidence. Candidates are then typically given a tour of the department, labs and university. They need to show interest, ask questions, and be enthusiastic. They should be prepared to meet the dean and the department head and to convince them also that they can deliver what they propose.

What Can Happen after the Interview

Even after successfully completing all the steps necessary to obtain the job, candidates may not be selected for the position, even if they thought their interview and reception were "perfect." Someone else may indeed have done something better to get the job, or perhaps one of the members of

the search committee, a powerful professor, simply didn't respond positively to a particular candidate. These are factors that are out of the candidate's control. Usually, if the search committee does not respond within a month, it is likely that they are negotiating with another candidate for the position and may be holding other candidates in reserve. Successful candidates therefore keep looking for a job until they have actually signed a contract. Candidates for any position should not accept a verbal offer as a final offer. Above all, they should not feel bad if they are rejected. It happens to almost everybody.

What to Do if Offered a Position

Nine-month starting salaries depend on the field. Starting salaries by field are listed by professional societies and other colleges. Deans hope to get faculty to work for as little as possible, even for choice faculty they consider their pot of gold at the end of the rainbow. Startup packages to support new faculty include funds (\$200,000 is common), facilities (labs, equipment, computers, etc.), salary for the summer, teaching relief (fewer classes assigned in first year or two), graduate student support from special sources, and equipment-matching funds. In most cases candidates negotiate their startup package. They need therefore to be smart and careful, which includes the teaching load they accept. They need to know, for example, if they will have teaching assistants, and whether or not they can "buy out" of a class in order to devote more time to research. Buying out may cost from 10 to 25% of annual salary. Various forms of taxation must also be taken into consideration. After all is said and done, the successful candidate may be the lucky one to find a faculty position that satisfies every expectation. 

COMBUSTION RESEARCH FACILITY NEWS

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